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Abstract

Organic-Inorganic Hybrid Mesoporous Silica Materials and their Application a
Matrix for Protein Molecules
Hua Dong
Dr. Yen Wei (Supervisor)

Transparent, organic-inorganic hybrid mesoporous silica materials hav(prepared successfully via the acid-catalyzed hydrolysis and cocondensation of tetramethyl orthosilicate and various organosiloxanes via the nonsurfactant tem| gel process. The organic groups were attached to the silica matrix via the non-hydrolyzable Si-C covalent bond and functioned as network modifier. The syntl conditions have been systematically studied and optimized. Nitrogen adsorptior transmission electronic microscopy characterizations show that the sol-gel matr obtained after removing the templates possess a three-dimensional network of interconnected mesopores. The pore parameters are tunable to some extent by v template content.

Several enzyme systems, such as horseradish peroxidase (HRP), glucos (GOx), lipase, alcohol dehydrogenase (ADH) etc. have been immobilized *in sitı* hybrid mesostructured, or more generally, nanostructured silica matrix. The cat: activity of immobilized enzymes has been assayed and correlated with the microstructures of the host silica materials under varied conditions. The enzyme encapsulated in the nonsurfactant-templated mesoporous sol-gel materials exhit

remarkably higher apparent catalytic activity, from a few-fold to three-orders of magnitude greater, than those in the non-templated conventional microporous h synthesized in the absence of the templates under otherwise identical conditions

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immobilization of more than one enzyme in the mesoporous host materials has l achieved, in which one enzyme's product is another's substrate. Protein unfoldi refolding in the nanoporous host have been investigated. Thermal stability of en were found to improve remarkably in the host materials.

Nonionic poly(ethylene oxide) copolymer surfactants were also utilizec templates for *in situ* immobilization of multiple enzymes. To minimize the dena of the enzyme in methanol which is liberated during the hydrolysis, a low-shrin gel process is also adapted to the direct immobilization of organophosphorus ac anhydrolase (OPAA). The thus immobilized OPAA showed high resistance to c solvent and is promising as the enzyme-based decontaminant. This study demoı that the novel sol-gel immobilization methods are versatile in terms of enzymes templates and matrix chemical compositions, leaving much room for further mc and optimization. The optically transparent, biologically doped sol-gel mesopor materials have potential applications in biocatalyst, biosensor devices, etc.

Preface

Since the discovery of highly ordered M41S family mesoporous molecı in 1992, considerable attention has been focused on tailoring the chemical comț these materials via the surfactant templated hydrothermal synthesis. Prior to thiṣ work, a novel nonsurfactant-templating pathway has been developed in our lab. of the template via water/solvent-extraction yields mesoporous silica materials ' surface area and pore volume as well as narrowly distributed pores of 2-6 nm. T

parameters can be tuned simply by varying the concentration of the templating compounds such as D-fructose, D-glucose, dibenzoyl-tartaric acid, malto cyclodextrin, oligopeptides, agar, etc. The ambient synthesis condition and biocompatibility of the template make direct immobilization of several enzyme: practical.

The objectives of this thesis study are to extend this nonsurfactant-temp pathway to the synthesis of optically transparent, hybrid organic-inorganic silic mesoporous materials and direct immobilization of biomolecules in such materi obtain a better understanding of the structure-property relationship, the activity immobilized enzymes is correlated with the pore sizes and functionalities of dif silica matrix. In Chapter 1, a brief review is given on the mesoporous materials application as the host matrix for the biomolecules immobilization. In Chapter 2 Chapter 3, a variety of organic-modified mesoporous silica materials based on t condensation of tetramethyl orthosilicate (TMOS) and organosiloxanes are desc The synthesis of hybrid mesoporous materials with controllable pore size and p

volume provides us with the means for direct immobilization of enzymes and o

biologically active species. In Chapter 4, we successfully applied this nonsurfac templated synthesis to the immobilization of horseradish peroxidase (HRP) in tl modified hybrid silica materials. The activities of the HRP encapsulated in the mesoporous hybrid hosts are significant higher than those in the non-templated microporous materials synthesized by the conventional sol-gel process. The the stability is also greatly enhanced because of the confinement in the nanometer-s cages. In Chapter 5, a multiple enzyme system, glucose oxidase and horseradish peroxidase, is co-immobilized in the pure and phenyl-modified mesoporous sili nonionic PEO surfactant is used as template under room temperature. The co-immobilized enzymes showed enhanced thermal stability and fast response to tl glucose substrate within few seconds.

In Chapter 6, organophosphorus acid anhydrolase (OPAA), an enzyme catalyze the hydrolysis of the toxic organophosphate, is immobilized in the mes silica matrix to develop a enzyme-based decontaminant in chemical warfare. Ar low-shrinking sol-gel process is employed to minimize the denaturation of the (methanol liberated during the sol-gel process. The immobilized OPAA has a hig remaining activity and resistance to organic solvents than free OPAA in solutio this novel templated immobilization is versatile in terms of enzymes, templates, chemical compositions of the silica matrix. The optically transparent, bioactive have numerous potential applications as biocatalyst and biosensor for target mo